Early Journal Content on JSTOR, Free to Anyone in the World

This article is one of nearly 500,000 scholarly works digitized and made freely available to everyone in the world by JSTOR.

Known as the Early Journal Content, this set of works include research articles, news, letters, and other writings published in more than 200 of the oldest leading academic journals. The works date from the mid-seventeenth to the early twentieth centuries.

We encourage people to read and share the Early Journal Content openly and to tell others that this resource exists. People may post this content online or redistribute in any way for non-commercial purposes.

Read more about Early Journal Content at http://about.jstor.org/participate-jstor/individuals/early-journal-content.

JSTOR is a digital library of academic journals, books, and primary source objects. JSTOR helps people discover, use, and build upon a wide range of content through a powerful research and teaching platform, and preserves this content for future generations. JSTOR is part of ITHAKA, a not-for-profit organization that also includes Ithaka S+R and Portico. For more information about JSTOR, please contact support@jstor.org.

CHLORINATED LIME AND HALAZONE IN THE DISINFECTION OF DRINKING WATER

BERNARD FANTUS

From the John McCormick Institute for Infectious Diseases, Chicago

The desirability of possessing, in a convenient and readily portable form, a reliable disinfectant for small quantities of water, such as might be carried in a soldier's drinking bottle, has led Dakin and Dunham[1] to advocate p-sulphondichloraminobenzoic acid ($Cl_2N.O_2S.C_6H_4.COOH$), under the name of halazone, as the best agent they were able to find, suitable for this purpose.

The present work was undertaken chiefly to compare the value of this new agent with that of chlorinated lime. To arrive at a practical conclusion, the following questions had to be answered regarding these two bodies: What is their relative germicidal power? What is their comparative toxicity? How do they compare in regard to suitableness for tablet-making (for a tablet is the ideal form for the purpose in mind)?

COMPARATIVE GERMICIDAL POWER

To ascertain this, I at first resorted to the determination of the phenol coefficient, according to the Hygienic Laboratory method.[2] A considerable amount of time devoted to this attempt, made it evident that the method was unsuitable for the purpose, on account of the following reasons: (1) This method arbitrarily fixes short time limits for action, 2½ and 15 minutes; when, in point of fact, the bodies studied developed their best action after the specified period. (2) It presupposes that there exists a simple and parallel relation of action according to concentration of solution and time of action. This, it has been found, does not exist in case of the two substances under consideration. (3) The medium, in which the disinfectant is permitted to act, is far too heavily polluted with bacteria (one-tenth ml. of 24-hour typhoid culture to 5 ml. of liquid), to make the determination of practical value for purposes of water disinfection. (4) The disin-

Received for publication Aug. 6, 1918.

Aided by a Grant from the Fenger Memorial Fund.

[1] Brit. Med. Jour., 1916, 1, p. 160; 1917, 1, p. 682, and 1917, 2, p. 790; also Am. Jour. Med. Sc., 1917, 154, p. 181.

[2] Anderson, J. F., and McClintic, T. B.: Hygienic Laboratory Bulletin, No. 82, Apr., 1912.

fectant is added to the medium before the bacteria are introduced. This causes an unfair destruction, by materials present in the medium, of such unstable chemicals as the chlorin bodies studied. In practical work the bacteria are present before the disinfectant is added.

The standard method of examining disinfectants proposed by the committee of the American Public Health Association[3] suffers from similar defects for the purpose in view. It must, of course, be recognized that this method is intended merely to serve for the preliminary classification of disinfectants; and that, for special purposes, special methods have to be devised.

To meet the requirements of the purpose in mind, the following modifications of the standard method were elaborated: No time limit was set to the experiment. On the contrary, the time was extended, until it became evident that further increase in time was useless, thus employing a natural rather than an arbitrary time limit. The results were charted on a logarithmic basis according to time and concentration, which permits showing at a glance the relationship of bodies differing so enormously in degree of action, as do these substances in comparison with phenol. A fairly large amount of water, 100 ml., was infected with 0.1 ml. of 24-hour broth culture of typhoid organisms, which gave a water pollution of about 20,000-30,000 per ml. The disinfectant was added after the organisms had been introduced.

Technic.—A 24-hour typhoid culture grown in standard extract broth, of a reaction of + 1.5 acid to phenolphthalein, was filtered through sterilized filter paper; and 0.1 ml. accurately measured by means of a Fournier syringe, discharged into 100 ml. of water contained in wide-mouthed sterile glass-stoppered bottles. These were brought to a temperature of 20 C., and kept at about this temperature by means of a water-bath like that described by Anderson and McClintic.[2] It was, however, difficult to maintain this temperature absolutely at this point in the long-time experiments during hot weather. At times, the temperature rose to 21 or 22 C. This may account for some of the variations in results. After a control culture had been taken, the accurately weighed or measured disinfectant was added; the contents of the bottle mixed by shaking, and subcultures taken at intervals, shorter in the higher concentrations and longer in the greater dilutions. The results were read after 48 hours' incubation by noting the turbidity of the tubes in which growth had taken place. Growth was recorded by + or ×; no growth by O. When tap water was used, growth was recorded by T; no growth by □

Inasmuch as chlorin is essentially the active agent in halazone as well as in chlorinated lime, it is of considerable theoretic as well as practical interest to determine whether the chlorin in the form of chloramin is as active as in that of the hypochlorite. Hence strictly equiatomic quantities were compared with each other. Thus an N/10 halazone powder was prepared in the following manner. A little more than 0.0675 gm. each of halazone and of anhydrous sodium carbonate were mixed with 9.865 gm. of sodium chlorid. This was titrated with

[3] Am. Jour. Publ. Health, 1918, 8, p. 506.

N/10 sodium thiosulphate and enough sodium chlorid added to make 1 gm. of the powder exactly equivalent to 1 ml. of the solution. In a similar manner, an N/10 powder of chlorinated lime was prepared by so adjusting the "available chlorin" in the mixture with sodium chlorid that each gm. would correspond to 1 ml. of N/10 sodium thiosulphate, when tested in the usual manner, by means of sodium iodid and acetic acid, using starch solution as an indicator. Each time before conducting a test, the powder was reassayed; and, whenever deterioration had occurred, allowance was made for it.

In plotting the results on cross-section paper, it became necessary to use a logarithmic scale, in order to bring the chart within bounds of convenience. In Charts 1 and 2, the concentration of the disinfectant has been indicated as ordinates on a logarithmic scale; and the time as the abscissas, likewise logarithmically. The figures at the left of the chart indicate fractions of N; the higher the figure the lower the concentration. The figures at the bottom of the charts indicate minutes. Only the crucial points of the various observations are noted; that is, a large number of + signs might have been placed to the left side of the one recorded, and a large number of zeros to the right of the O shown in the table, if all the tests made had been noted on the chart. For the sake of clearness, this has not been done. Where two or more experiments did not quite correspond, this is indicated. A few great aberrations from the average results were discarded, as in some of these error of technic was discovered; in others, such error was probable even though not discovered. The line or curve expressing the action of the agent was drawn as nearly as possible midway between the + and O signs, but so as to leave few or no + signs to the right of the line. The position on the table of a line thus drawn, or rather a zone to each side of it, may be taken to represent the activity of the drug. It is approximately correct not only for the concentration tested, but also for intermediate time and concentrations, as has become evident over and over again. The relative position of these lines to each other may be taken to represent the relative activity of the substances tested.

The following signs are used on Charts 1 and 2 to distinguish graphically between the various data to be shown:

+ = Growth—disinfectant acting in distilled water.
O = No growth—disinfectant acting in distilled water.
⊕ = Growth and no growth in two different experiments.
T = Growth in tap water.
□ = No growth in tap water.
× = Growth after potassium permanganate in distilled water.
◇ = No growth after potassium permanganate in distilled water.
— = Line of action in distilled water.
.... = Line of action in tap water.

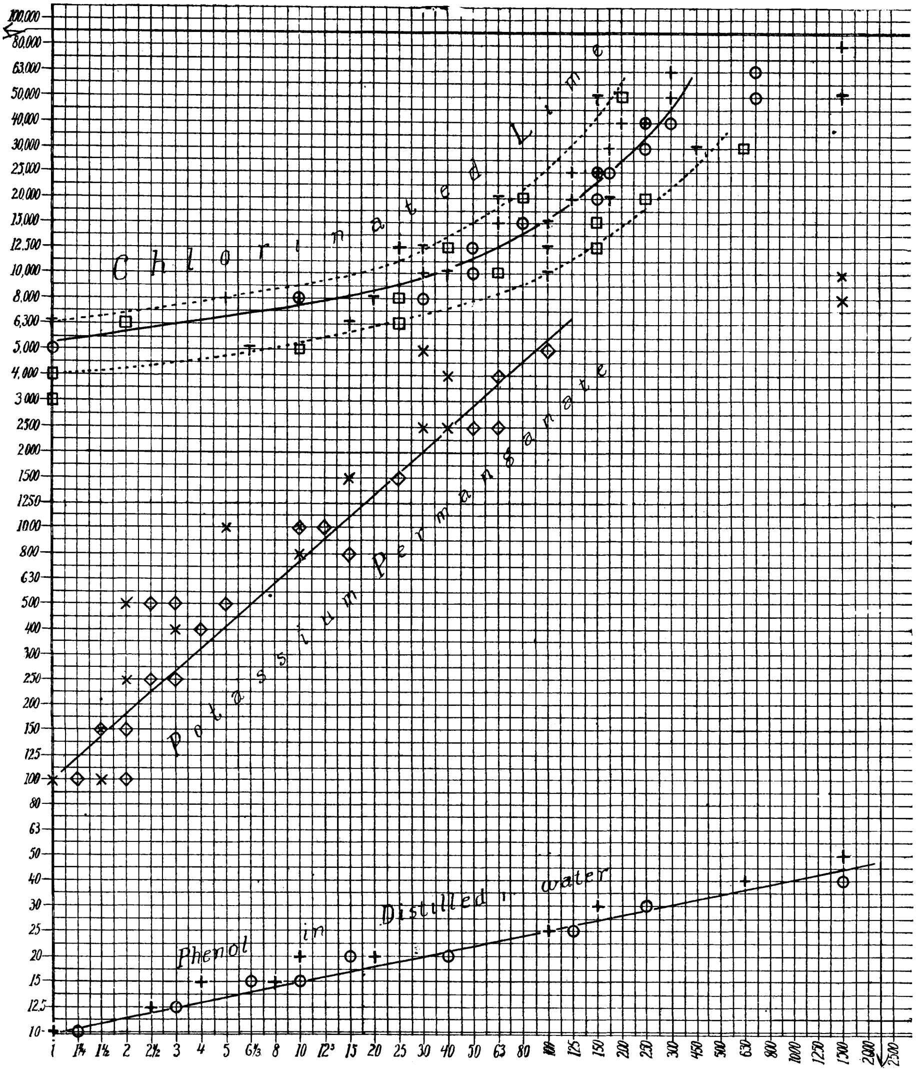

Chart 1.—Record of the experiments made with phenol, chlorinated lime, and permanganate.

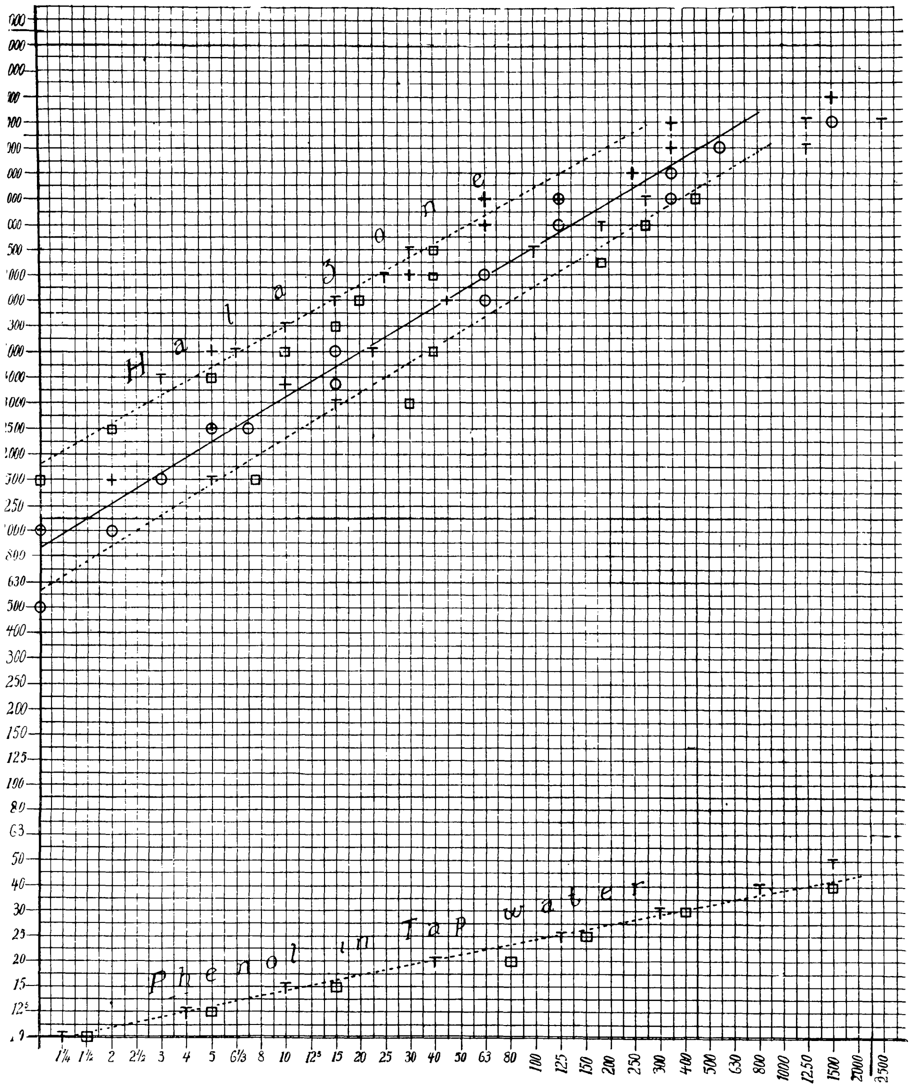

Chart 2.—Comparative results of the experiments made with halazone and phenol.

It will be noted by reference to Chart 1 that the activity of phenol ranges from N/10 (approximately 1:100), which sterilizes infected distilled water in 1¼ minutes, to N/40, which requires about 24 hours for sterilization. A solution of N/50 (1:500) phenol does not sterilize even after 24 hours. At the bottom of Chart 2 is shown the result of an experiment on the action of phenol in tap water. The line is parallel to that of phenol in distilled water and merely shifted somewhat to the right, showing that a slightly longer time is required for sterilization. Thus, with N/10 phenol it was 1½ minutes instead of 1¼; with N/15, 15 minutes were needed in tap water, as compared with 10 minutes in distilled water. It will be seen at a glance that the disinfecting power of phenol decreases rapidly with decrease in concentration.

To obtain sterilization within 1 minute, N/5,000 of chlorinated lime would have to be used. This equals approximately 7 parts of available chlorin per million. From here the unbroken line (Chart 1), showing the action of this agent in distilled water, runs almost straight to N/10,000 which requires, in our experiment, 50 minutes for sterilization of infected distilled water. From this point a change in the direction of the line takes place, so that it becomes a curve, which ends at N/63,000. A solution as dilute as N/80,000, fails to disinfect even after 24 hours. The reason for the curving of the line I am unable to give at this time. That it is neither an accident nor an artefact, that is, due to rise of temperature, I feel able to assert on the strength of sufficiently repeated and, at times, sufficiently carefully controlled experiments. It may be that the relative improvement in the rate of action with greater dilutions is due to greater ionization in the more dilute solution. When Chicago tap water infected with typhoid bacilli was used, the results were much less consistent than with distilled water; at times, the disinfection occurred more rapidly in tap water than in distilled water; at times, more slowly, as shown by the dotted lines on Chart 1. This is evidently due to variability in the composition of the Chicago water supply, due to the effect of prevailing wind and weather on the water of Lake Michigan, near the shore. The rather surprising fact that, at times, the chlorinated lime acted more rapidly in tap water than it did in distilled water, might be due to the presence of a greater number of H-ions, which have an accelerating action, as shall be shown, or may depend on the fact that from 3-5 lbs. of liquefied chlorin are added to each million gallons of water at the Chicago water works.

The line representing the action of halazone (Chart 2) starts at N/1,000 (calculated as "available chlorin"), approximately 1:15,000 of halazone by weight. This disinfects within 2 minutes. The line extends to N/40,000, which requires almost 24 hours for action. At N/50,000 the action ends. It should be noted, that the line is straight. This would make the theory, that the curve in the chlorinated lime line is due to ionization, agree best with the experimental facts, as halazone does not ionize. With tap water, variable results were obtained, as in case of chlorinated lime. At times, the action was

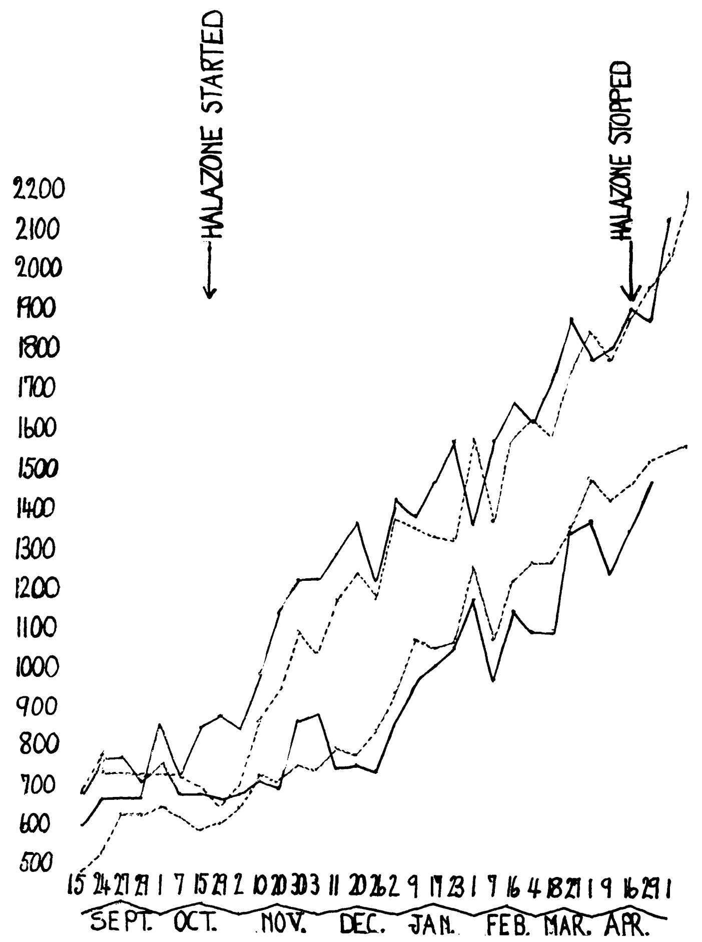

Chart 3.—Weight curve of cats given 0.1 gm. halazone daily with their food (continuous line), compared with weight curve of control animals (broken line).

slower than in distilled water; at times, it was more rapid. Dakin and Dunham[1] found that 1:300,000 of halazone disinfected heavily polluted water within 30 minutes. This approximately equals N/20,000 halazone, which in our experiments required at an average 200 minutes in distilled water; though, in tap water, speedier as well as slower action might be obtained. This makes halazone, in the concentration recommended, appear to be a rather slow disinfectant.

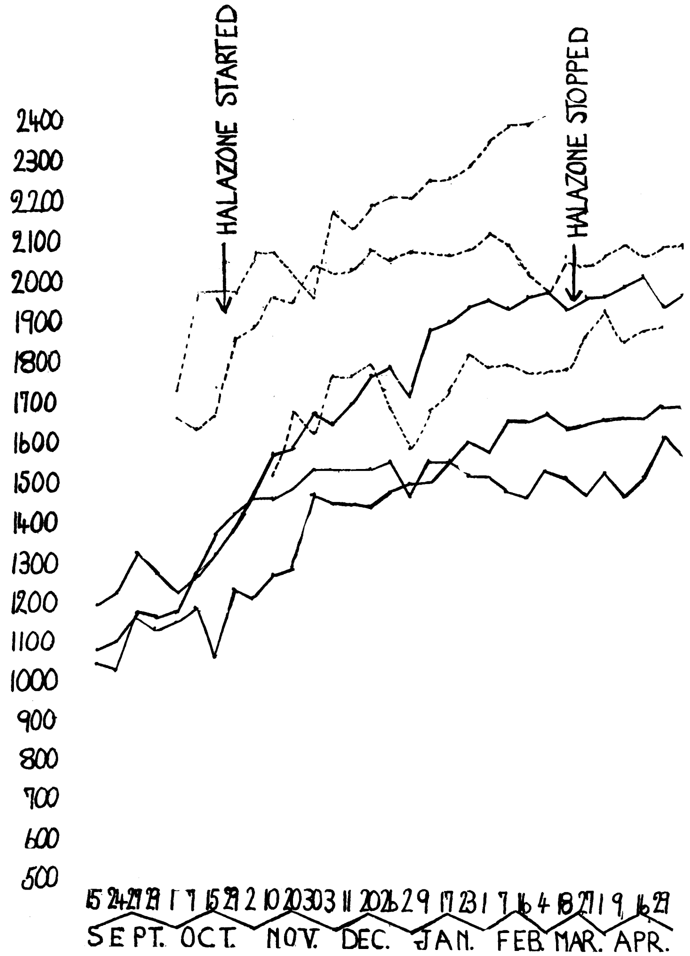

Chart 4.—Weight curve of rabbits given 0.1 gm. halazone daily (continuous line), compared with that of control animals (broken line).

To compare chlorinated lime with halazone in terms of the phenol coefficient at 20 C. for 5-20 minutes, the relation would be as follows:

Chlorinated lime	466
Halazone	228

In other words, compared on the basis of available chlorin, chlorinated lime is about twice as efficient a water disinfectant as is halazone. This superior activity is evidently due to the greater chemical instability of the chlorinated lime.

DETERMINATION OF TOXICITY

While it is well known that the products remaining after chlorinated lime has exerted its action in water disinfection are harmless, it seemed of interest to determine the toxicity of the decomposition products of halazone, most especially when fed daily for a considerable length of time. Hence, feeding experiments were undertaken, which may be described as follows:

Feeding Exper. 1.—Four kittens of one litter, of approximatey similar weight, which at the beginning of the experiment ranged from 500-700 gm., were kept in separate cages and fed weighed quantities of food, consisting of milk mixed with cornmeal mush. Later, meat was given on alternate days. The quantity of food was so adjusted that the animals would empty their dishes; and, of course, was uniform for all 4 animals. After a preliminary period of 6 weeks, 2 of the animals were given halazone, while 2 were kept as controls. For medication of the food, a mixture of 0.1 gm. of halazone and 0.1 gm. of anhydrous sodium carbonate was added daily to the food. The animals consumed this medicated food almost as well as did the control animals, and gained in weight to practically the same extent as the controls, as will be seen from Chart 3, which shows the weight curve of the control animals in broken lines and of the animals that received the medicated food, in continuous lines. It will be noted that the lines are practically parallel and that the control animals did no better than those that were given the halazone mixed with their food. The animals showed no symptoms of intolerance. At the conclusion of the experiment, the halazone animals were chloroformed and their organs examined. There was no macroscopic evidence of damage to any of the organs usually examined at necropsy. Microscopic examination of the livers and kidneys showed no changes.

Feeding Exper. 2.—Three rabbits, of approximately equal weight (1050-1225 gm.), were kept in separate cages; and fed with chopped carrots mixed with oats, in such quantity as to insure complete consumption by the animal during 24 hours. After a control period of about 1½ months, the animals were given 0.1 gm. halazone and 0.1 gm. anhydrous sodium carbonate daily, mixed with their food, which they consumed quite as readily as they did the unmedicated food previously administered. Three control animals, also kept in separate cages, were fed the same amount of oats and carrots. Chart 4, which illustrates the result of this experiment, shows that the gain in weight of all the 6 animals was sufficienty parallel to enable one to say that the halazone addition to the food evidently did not interfere with the growth and development of the rabbits that received it. The weight of 2 of the control rabbits was excessive at the beginning of the experiment, as compared with the weight of the others; the rate of their gain in weight, however, was quite proportional to that of the halazone animals. The necropsy of the halazone animals showed nothing abnormal.

Exper. 3.—Acute Poisoning. Cat 1, weight 2,700 gm., was given 2.7 gm. each of halazone and sodium carbonate dissolved in water (1 gm. per kg.) by stomach tube. Almost immediate and repeated profuse emesis was the only symptom noted. The animal recovered.

Cat 2, weighing 3,200 gm., was given 6.4 gm. of halazone (2 gm. per kg.) by means of a stomach tube. Profuse emesis, setting in within 4 minutes and repeated 3 times during the course of the next few hours, was the only result. The animal recovered.

Cat 4, weight 1,600 gm. was given 3.2 gm. of halazone (2 gm. per kg.) per stomach tube. Emesis set in 10 minutes later, and continued at intervals throughout the day. At first the vomitus consisted of food mixed with the injected material, later it was mucous. The animal died on the 2nd day after the administration of the dose; and showed, on necropsy, evidence of violent irritation of the stomach. Otherwise the organs seemed negative, though microscopic examination was not made, as the necropsy was performed too late to obtain satisfactory results from such examination.

DISCUSSION OF THE FEEDING EXPERIMENTS

The fact that the halazone was administered mixed with the food in the feeding experiments makes it evident that the chlorin was to a great extent lost by decomposition of the chloramin before the animal could eat the food. It was, therefore, chiefly p-sulphonaminobenzoic acid that the animals ingested. This probably accounts for the lack of irritation of the stomach from consumption of this food. Inasmuch as the sulphondichloraminobenzoic acid (halazone), when used for water disinfection, is almost wholly decomposed in a similar manner before the disinfected water is ingested, the feeding experiments may be considered to show that such water could not have any deleterious effects on the human organism, especially when it is realized that 0.1 gm. per kg. would be equivalent to the ingestion of 6 gm. of halazone by a man weighing 60 kg. Such quantity of halazone would be sufficient to disinfect 600 liters of polluted water.

The experiments on acute poisoning show that large doses of halazone, whether taken in suspension or in solution by the aid of sodium carbonate, acts as a violent gastric irritant, which by reason of the prompt emesis that it provokes would hardly be likely to produce death in a human being.

TABLET-MAKING QUALITIES

Inasmuch as for the disinfection of small quantities of drinking water, such as those that might be gathered and carried by rapidly moving troops, tablets constitute by far the most satisfactory form for use, the study of the suitableness of these agents for tablet-making becomes of special importance.

As lime in the form of chlorinated lime is the cheapest vehicle for chlorin and it proved itself in our studies the most efficient disinfectant, the question of the preparation of chlorinated lime tablets was taken up. I am indebted to Dr. F. O. Tonney, director of the Municipal Laboratories of the Chicago Health Department, for permission to publish the data obtained by Mr. Jay Kaplan in an inquiry, taken up in 1916, for the purpose of devising water disinfecting tablets.

The tablets were prepared by slightly moistening chlorinated lime in a mortar to make a thick paste. This was pressed in the perforations of a tablet triturate mold of vulcanite. After drying by placing the mold for from 5-10 minutes in an oven at 40-50 C., the tablets were carefully forced out by fitting the 2 parts of the mold together and applying slow pressure. The tablet triturates thus prepared were shaken into bottle and tightly stoppered. Tests showed that, under average conditions, the disinfectant retained its potency for about 4 weeks. In four months a deterioration of about 40% had occurred. Seven and one-half months later, the tablets were found to be moist and sticky so that they did not retain their shape; and they had undergone a deterioration of 82.5%. Hence, it may be concluded that chlorinated lime tablets, prepared by the tablet triturate process, do not possess satisfactory keeping qualities.

On the assumption that the moisture employed in the preparation of the tablet triturates just described was responsible for the rapid deterioration of the tablets, I had some tablets prepared by means of compression, using the following formula:

Chlorinated lime, 30%, or proportionately larger amounts of weaker lime, 0.59 gm. Sodium chlorid (granular), 10.0 gm. Divide into 100 tablets weighing 105 mg., avoiding use of lubricant.

One of these tablets will disinfect 1 liter of moderately infected water within 1 hour in cool, or much sooner in hot weather.

These tablets, made May 3, 1918, without special care as to drying, and kept in the dark in a well stoppered bottle, were assayed from time to time with the following results:

May 6, 1918: 5 tablets weighing 0.538 gm. yielded 9.2 mg. available Cl.

June 15, 1918: 5 tablets weighing 0.544 gm. yielded 8.1 mg. available Cl.

July 8, 1918: 5 tablets weighing 0.529 gm. yielded 7.1 mg. available Cl.

Aug. 9, 1918, 5 tablets weighing 0.536 gm. yielded 6.5 mg. available Cl.

Sept. 6, 1918: 5 tablets weighing 0.533 gm. yielded 5 mg. available Cl.

This is equivalent to a deterioration of about 10% per month. Had the tablets been prepared from dried material, the deterioration would, no doubt, have been slower. Nevertheless, it must be admitted that chlorinated lime tablets are decidedly unstable. However, by the time the deterioration has reached 50%, which might be in the course of half a year, these tablets would still be as good as ever for disinfecting half the quantity of water.

It is, indeed, this very unstability of chlorinated lime tablets that is the raison d'étre for halazone. Dakin and Dunham propose the following formula for these tablets:

Halazone	4 gm.
Sodium carbonate (dried)........	4 gm.
Sodium chlorid	92 gm.

The material should be carefully dried and mixed, the alkali being added last, and made into 100 mg. tablets, without use of lubricant. They claim that 1 such tablet is capable of disinfecting 1 liter of water in 30-60 minutes. In our experiments, still longer time was required for disinfection.

Commercial halazone tablets showed the following results on assay from time to time:

Bottle opened May 6, 1918.

May 6: 5 tablets, weighing 0.550 yield 10.6 mg. available Cl.
July 8: 5 tablets, weighing 0.561, yield 10.6 mg. available Cl.
Aug. 9: 5 tablets, weighing 0.573, yield 10.6 mg. available Cl.
Sept. 6: 5 tablets, weighing 0.577, yield 11.2 mg. available Cl.

Therefore, there had been practically no deterioration in six months.

CONCLUSIONS

Chlorinated lime is a more efficient water disinfectant than halazone, compared on the basis of active chlorin. It is, of course, also a great deal cheaper. Hence there is no reason for employing halazone in water disinfection, excepting when its superior tablet-making qualities render its use advantageous.

Halazone is the better agent for the preparation of water disinfecting tablets. Its action is, however, a rather slow one.